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(54) [Title of the Invention]

MULTILAYER-CIRCUIT SUBSTRATES AND A METHOD FOR THEIR PRODUCTION

(57) [Abstract]

[Problem to be Solved] To provide a method for the production of multi-layer-circuit substrates with good appearance, sufficient dimensional stability, and high adhesive strength at low cost, and the multilayer-circuit substrates that can be obtained by said method.

[Solution] Method for the production of multilayer-circuit substrates with a thermoplastic liquid-crystal polymer film as an insulating material, wherein a thermoplastic liquid-crystal polymer film 1 with high heat resistance and a metal sheet 2 are placed one over the other and pressure bonded with a pair of heating rolls 3, then a circuit is formed on this metal sheet 2 to prepare a circuit substrate 5 with high heat resistance, and subsequently this circuit substrate 5 with high heat resistance is arranged on the upper and lower surfaces of a thermoplastic liquid-crystal polymer film 6 that has lower heat resistance than the thermoplastic liquid-crystal polymer film 5 with high heat resistance, and pressure bonded with a pair of heating rolls, 7, 7.

[Scope of the Patent Claims]

[Claim 1] Method for the production of multilayer-circuit substrates, which is a method for the production of multilayer-circuit substrates that have a film (hereinafter called a thermoplastic liquid-crystal polymer film) consisting of a thermoplastic polymer that can form an optically anisotropic molten phase, as an insulating material, and characterized in that a thermoplastic liquid-crystal polymer film with high heat resistance and a metal sheet are placed one over the other and pressure bonded with a pair of heating rolls; in that a circuit is then formed on the metal sheet to prepare a circuit substrate with high heat resistance; and in that the obtained circuit substrate with high heat resistance is then arranged on the upper and lower surfaces of a thermoplastic liquid-crystal polymer film having lower heat re-

sistance than said thermoplastic liquid-crystal polymer film with high heat resistance, and pressure bonded with a pair of heating rolls.

**[Claim 2]** Method for the production of multilayer-circuit substrates, which is a method for the production of multilayer-circuit substrates with a thermoplastic liquid-crystal polymer film as an insulating material, and characterized in that a thermoplastic liquid-crystal polymer film with high heat resistance and a metal sheet are placed one over the other and pressure bonded with a pair of heating rolls; in that a circuit is then formed on said metal sheet to prepare a circuit substrate with high heat resistance; in that the obtained circuit substrate with high heat resistance is then arranged on the upper and lower surfaces of a thermoplastic liquid-crystal polymer film having lower heat resistance than said thermoplastic liquid-crystal polymer film with high heat resistance, and pressure bonded with a pair of first heating rolls; and in that further said thermoplastic liquid-crystal polymer film with lower heat resistance is placed over the upper and lower surfaces of the obtained multilayer-circuit substrate and pressure bonded with a pair of second heating rolls.

**[Claim 3]** Method for the production of multilayer-circuit substrates, wherein the change in the dimensions of said circuit substrates before and after pressure bonding with the heating rolls is 0.1% or less and the adhesive strength between said circuit substrate and thermoplastic liquid-crystal polymer film with lower heat resistance is 0.5 kg/cm or more in Claim 1 or Claim 2.

**[Claim 4]** Multilayer-circuit substrates that can be obtained by the production method described in Claim 1 or Claim 2.

**[Detailed Description of the Invention]**

**[0001]**

**[Technical Field of the Invention]** The present invention pertains to a

method for the continuous production of multilayer-circuit substrates using a film (hereinafter may be called a thermoplastic liquid-crystal polymer film) consisting of a thermoplastic polymer that can form an optically anisotropic molten phase, and to multilayer-circuit substrates that can be obtained by said method. The multilayer-circuit substrates of the present invention have excellent dimensional stability, low moisture absorption, heat resistance, chemical resistance, and electrical properties arising from the thermoplastic liquid-crystal polymer film used as the electrically insulating material, and are useful as a base material for flexible circuit boards and semiconductor packaging circuit substances.

[0002]

**[Conventional Techniques and Problems to be Solved by the Invention]** In producing a multilayer-circuit substrate to be used in printed circuit boards, etc., by using a thermoplastic liquid-crystal polymer film, the multilayer-circuit substrate has hitherto been obtained by using a vacuum hot press system, placing a thermoplastic liquid-crystal polymer film cut to a prescribed size and a metal sheet one over the other between two hot flat plates of the system, bonding them by heat and pressure in a vacuum condition (batch-mode vacuum hot press lamination process), then forming a circuit on the metal sheet, to prepare a circuit substrate, then heat and pressure bonding this substrate with a thermoplastic liquid-crystal polymer film with different heat resistance in a vacuum condition. However, the vacuum hot press lamination process is a sheet-fed process and hence there is a problem in that the time to stack the materials, one pressing time, and the time to remove the material after pressing are long, which reduces the production speed and leads to high costs. Furthermore, if the facility is improved so as to be able to produce a large number of substrates at the same time in order to increase the production speed, the facility becomes large and the cost of said facility in-

creases, which is undesirable. What is more, mismatches occur between the metal sheets and the thermoplastic liquid-crystal polymer sheets, or between the circuit substrate and the thermoplastic liquid-crystal polymer sheets during press bonding by heat and pressure, or the resin may flow to the circumference, which may detract from the appearance of the product. Accordingly, there is a need for a production method that eliminates these problems and that can provide multilayer-circuit substrates at low cost.

[0003] Furthermore, multilayer-circuit substrates are produced by using sheets that consist of materials with different chemical compositions and heat resistance (melting points), forming a wiring pattern (circuit) on the sheets, combining them, and then bonding them with heat and pressure. At that time, if the materials of the individual sheets of the multilayer-circuit substrates have the same melting point, both sheets will melt during heat and pressure bonding; accordingly, the melting point of one material must be made higher than the melting point of the other material (refer to Japanese Patent Kokai No. 8[1996]-97,565). Moreover, materials with different melting points usually have different chemical compositions, and adhesive strength is usually low at the interface between such materials. Therefore, if materials with different melting points are used, there are problems with peeling due to the process of preparing the multilayer-circuit substrate, or there are environmental changes after the product is made. Furthermore, there is a problem from the standpoint of the environment and resource management when the multilayer-circuit substrate is to be discarded after use, because it cannot be recycled effectively unless it is separated into individual materials.

[0004] Thermoplastic liquid-crystal polymer films have excellent properties such as heat resistance and chemical resistance. In preparing a multilayer-circuit substrate with the use of such films, the problem from the standpoint of the environment and resource management can be eliminated, since

the same materials are used. However, problems similar to those mentioned above occur even when using said films in the multilayer-circuit substrate; thus, these too must be eliminated. In other words, various heat-resistant materials with different melting points must be used to produce the multi-layer-circuit substrate, and moreover the dimensions of the substrate on which a circuit has been formed must not change excessively during the process of heat and pressure bonding with an adhesive sheet with lower heat resistance.

[0005] In accordance with the above, the object of the present invention is to provide a method for the production of multilayer-circuit substrates with good appearance, sufficient dimensional stability, and high adhesive strength at low cost, and the multilayer-circuit substrates that can be obtained by said method.

[0006]

**[An Approach to Solving the Problems]** To achieve the above-mentioned objective, the first invention of the present application pertains to a method for producing a multilayer-circuit substrate which uses a thermoplastic liquid-crystal polymer film as an insulating material, and consists of placing a thermoplastic liquid-crystal polymer film with high heat resistance and a metal sheet one over the other and pressure bonding these with a pair of heating rolls, subsequently forming a circuit on the metal sheet, to prepare a circuit substrate with high heat resistance, and subsequently arranging the obtained circuit substrate with high heat resistance on the upper and lower surfaces of a thermoplastic liquid-crystal polymer film with lower heat resistance than the foregoing thermoplastic liquid-crystal polymer film with high heat resistance and pressure bonding with a pair of heating rolls.

[0007] Furthermore, the second invention of the present application consists of further placing the foregoing thermoplastic liquid-crystal polymer film with lower heat resistance on the upper and lower surfaces of the multi-

layer-circuit substrate that can be obtained by the first invention, and pressure bonding with a pair of heating rolls.

[0008] The above-mentioned first and second inventions can be used for the production of multilayer-circuit substrates with good appearance with no resin flow to the circumference, sufficient dimensional stability and high adhesive strength at low cost. Furthermore, the second invention can be used for the production of a multilayer-circuit substrate with better dimensional stability, etc., because every part of the circuit formed on the circuit substrate is covered with the thermoplastic liquid-crystal polymer film.

[0009] In each of the above-mentioned inventions, the dimensional change in said multilayer-circuit substrate before and after pressure bonding with the heating rolls is preferably 0.1% or less, and the adhesive strength between said circuit substrate and thermoplastic liquid-crystal polymer film with lower heat resistance is preferably 0.5 kg/cm or more. According to this arrangement, a multilayer-circuit substrate with better dimensional stability as well as good appearance and higher adhesive strength can be obtained. If said dimensional change exceeds 0.1% or the adhesive strength is less than 0.5 kg/cm, the dimensional stability will be unsuitable, and, moreover, there will be variations in the multilayer-circuit substrate produced, which leads to insufficient adhesive strength and poor appearance.

[0010] There are no particular limitations with regard to the thermoplastic liquid-crystal polymer film to be used in the present invention, but specific examples include well-known thermotropic liquid-crystal polyesters and thermotropic liquid-crystal polyesteramides, which can be derived from the compounds classified as (1) to (4) below and their derivatives. However, it goes without saying that there are suitable limits to combinations of various starting compounds in order to obtain polymers that can form an optically anisotropic molten phase.

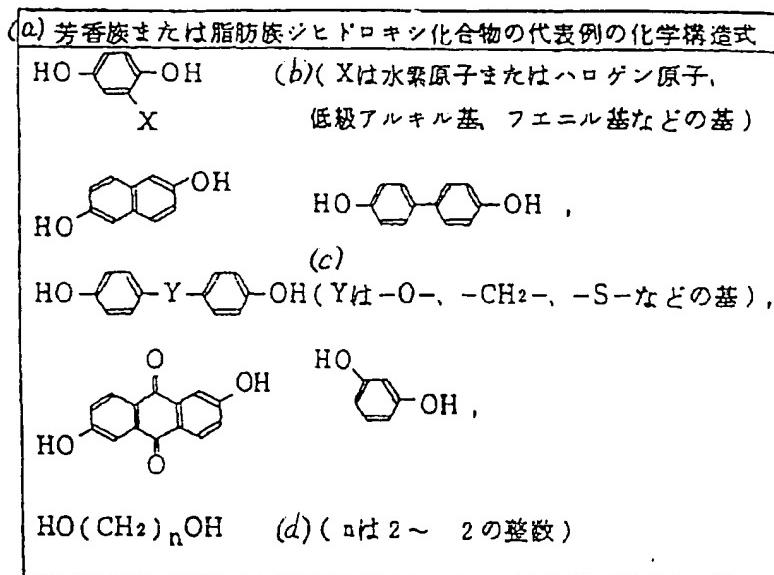
[0011]

(1) Aromatic or Aliphatic Dihydroxy Compounds (refer to Table 1 for typical examples)

[0012]

[Table 1]

TABLE 1. KEY: (a) chemical structural formulas of typical examples of aromatic or aliphatic dihydroxy compounds; (b) (X is a hydrogen atom, a halogen atom, or a group such as lower alkyl or phenyl); (c) (Y is a group such as -O-, -CH<sub>2</sub>, or -S-); and (d) (n is an integer of 2-2) [sic -- Tr. Ed.].



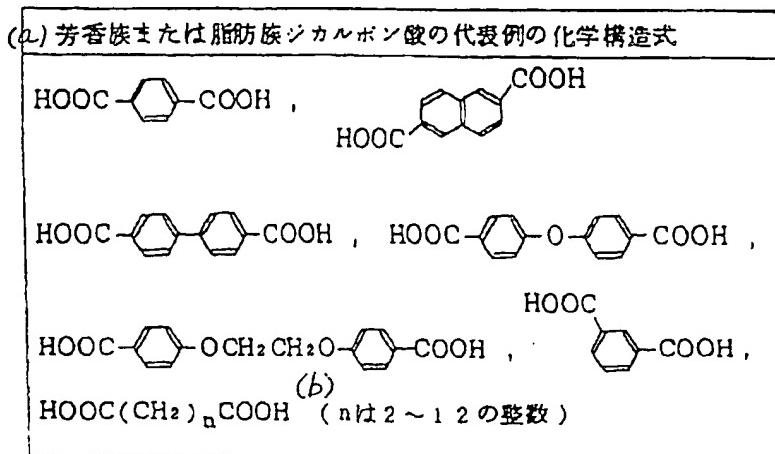
[0013]

(2) Aromatic or Aliphatic Dicarboxylic Acids (refer to Table 2 for typical examples)

[0014]

[Table 2]

TABLE 2. KEY: (a) chemical structural formulas of typical examples of aromatic or aliphatic dicarboxylic acids; and (b) (n is an integer of 2-12).



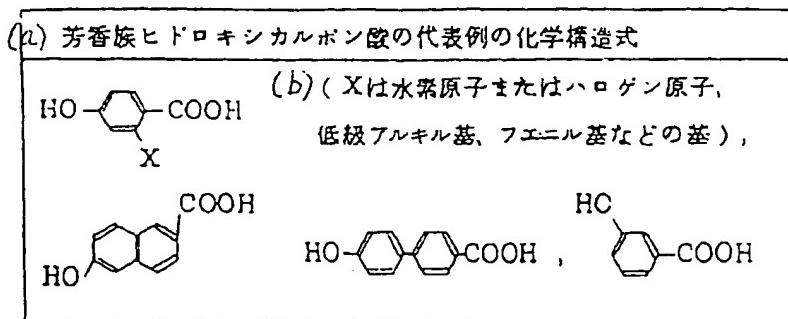
[0015]

(3) Aromatic Hydroxycarboxylic Acids (refer to Table 3 for typical examples)

[0016]

[Table 3]

TABLE 3. KEY: (a) chemical structural formulas of typical examples of aromatic hydroxycarboxylic acids; and (b) (X is a hydrogen atom, a halogen atom, or a group such as lower alkyl or phenyl).



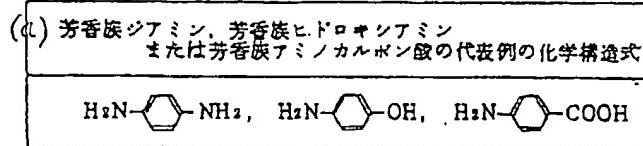
[0017]

(4) Aromatic Diamines, Aromatic Hydroxyamines, or Aromatic Aminocarboxylic Acids (refer to Table 4 for typical examples).

[0018]

[Table 4]

TABLE 4. KEY: (a) chemical structural formulas of typical examples of aromatic diamines, aromatic hydroxyamines, or aromatic aminocarboxylic acids.



[0019] Typical examples of thermoplastic liquid-crystal polymers that can be obtained from these starting compounds include copolymers (a)-(e) with the structural units shown in Table 5.

[0020]

[Table 5]

TABLE 5. KEY: (A) typical examples of thermoplastic liquid-crystal polymers; (B) copolymer; and (C) (X is a group such as -O-, -CH<sub>2</sub>- or -S-).

(A) 热可塑性液晶ポリマーの代表例	
(a)	$\text{---OC}(\text{---C}_6\text{H}_4\text{---CO})_n\text{---OCH}_2\text{CH}_2\text{O---OC}(\text{---C}_6\text{H}_4\text{---O---})_m\text{---O}$ 共重合体
(b)	$\text{---O}(\text{---C}_6\text{H}_4\text{---CO})_n\text{---O---}\left[\begin{array}{c} \text{---C}_6\text{H}_4\text{---CO---} \\   \\ \text{---C}_6\text{H}_4\text{---} \end{array}\right]_m\text{---O---}$ (B) 共重合体
(c)	$\text{---O}(\text{---C}_6\text{H}_4\text{---CO})_n\text{---OC}(\text{---C}_6\text{H}_4\text{---CO})_m\text{---O---}\left[\begin{array}{c} \text{---OC}(\text{---C}_6\text{H}_4\text{---CO})_n\text{---O---} \\   \\ \text{---C}_6\text{H}_4\text{---} \end{array}\right]_p\text{---O---}$ (B) 共重合体
(d)	$\left[\begin{array}{c} \text{---C}_6\text{H}_4\text{---CO---} \\   \\ \text{---C}_6\text{H}_4\text{---} \end{array}\right]_q\text{---OC}(\text{---C}_6\text{H}_4\text{---CO})_n\text{---O---}\left[\begin{array}{c} \text{---O---} \\   \\ \text{---C}_6\text{H}_4\text{---NH---} \end{array}\right]_r\text{---O---}$ (B) 共重合体
(e)	$\text{---O}(\text{---C}_6\text{H}_4\text{---CO})_n\text{---OC}(\text{---C}_6\text{H}_4\text{---CO})_m\text{---O---}\left[\begin{array}{c} \text{---O---} \\   \\ \text{---C}_6\text{H}_4\text{---X---} \end{array}\right]_p\text{---O---}$ (B) 共重合体 (C) (Xは-O-, -CH <sub>2</sub> -, -S-などの基)

[0021] Furthermore, thermoplastic liquid-crystal polymers with melting points in the range of about 200 to about 400°C, especially in the range of about 250 to 350°C, are preferred in order to achieve the objective of obtaining the desired heat resistance and workability of the film.

[0022] The thermoplastic liquid-crystal polymer film to be used in the present invention can be obtained by extrusion molding a thermoplastic liquid-crystal polymer. Any of the extrusion molding processes can be employed, but the well-known T-die process, laminate drawing process, and inflation process are industrially profitable. In particular, the inflation process and laminate drawing process can provide films that are well-balanced in mechanical

properties and thermal properties in the machine axis direction (hereinafter abbreviated as MD direction) of the film and in the direction orthogonal thereto (hereinafter abbreviated as TD direction), because of applying stress not only in the MD direction but also in the TD direction.

[0023] The material of the metal sheet to be used in the present invention can be selected from among metals that are used in electrical connections, and suitable examples include gold, silver, copper, nickel, and aluminum. Of these, copper is especially preferred. Copper that is produced by either a rolling process or the electrolysis process can be used, but the copper with large surface roughness produced by the electrolysis process is preferred. The metal sheet may be subjected to chemical surface treatment such as the acid pickling usually done for copper foil, without thereby impairing the effect displayed by the present invention. The thickness of the metal sheet is preferably in the range of 7-100  $\mu\text{m}$ , and especially in the range of 9-75  $\mu\text{m}$ .

[0024]

**[Embodiments of the Invention]** One embodiment of the production method of the present invention will now be explained with reference to the figures. Figure 1 is a diagram that schematically illustrates one example of producing a circuit substrate that can be used in the present invention. As shown in the figure, a two-side metal-lined laminate 4 with high heat resistance is prepared by pressure bonding a thermoplastic liquid-crystal polymer film 1 with high heat resistance and metal sheets 2, 2 by passing them between a pair of heating rolls 3, 3, upper and lower, while feeding said metal sheets 2, 2 so as to be stacked on the upper and lower surfaces of said film 1. And the laminate 4 is subjected to a treatment similar to that of the conventional circuit pattern preparation method, to prepare a circuit substrate 5. In other words, a photosensitive resin is applied or a photosensitive film is

pressure bonded on both the upper and lower metal sheet 2 of the laminate 4, a predetermined circuit pattern exposure is carried out, the resin other than the photosensitive resin circuit pattern is removed, and then etching is carried out to obtain the circuit substrate 5.

[0025] Figure 2 is a diagram that schematically illustrates one embodiment of the method for producing a multilayer circuit substrate of the present invention. The figure shows a method for producing a four-layer-circuit substrate, i.e., a circuit substrate wherein four circuits are provided in the thickness direction. In the figure, while feeding two pieces of the high-heat-resistance circuit substrates 5 obtained by the above-mentioned process onto the two surfaces, upper and lower, of a thermoplastic liquid-crystal polymer film 6 with a melting point lower than that of the aforesaid high-heat-resistance thermoplastic liquid-crystal polymer film 1, i.e., with lower heat resistance, they are passed between a pair of heating rolls 7, 7, upper and lower. In this way, each of these circuit substrates 5 and thermoplastic liquid-crystal polymer films 6 with lower heat resistance are pressure bonded into one integrated piece, to prepare a four-layer-circuit substrate A (first invention). To prevent damaging of the circuit formed on the outer surface of each circuit substrate 5 by each heating roll 7 in the above-mentioned process, a mold-release sheet 8 is placed around each heating roll 7 and rolls (not shown) arranged opposite, and each of these mold-release sheets 8 is brought into contact with the surface of said circuit substrate 5, to carry out pressing by each of the heating rolls 7.

[0026] Subsequently, two sheets of thermoplastic liquid-crystal polymer films 9, 9 with lower heat resistance are fed in at the front of the forward-going direction of the aforesaid first heating rolls 7 so as to sandwich the two surfaces, upper and lower, of the aforesaid four-layer-circuit substrate A, and these are passed between a pair of second heating rolls 10, 10, upper and

lower. In this way, said four-layer-circuit substrate A and thermoplastic liquid-crystal polymer films 9, 9 with lower heat resistance are pressure bonded into one integrated piece, to prepare a four-layer-circuit substrate B with both outer surfaces of the four-layer-circuit substrate A covered with thermoplastic liquid-crystal polymer film 9 with lower heat resistance (second invention). In this case also, a mold-release sheet 8 is placed around each heating roll 10.

[0027] The surface temperature of the above-mentioned first and second heating rolls 7, 10 is preferably in the range of a temperature that is 50°C lower than the melting point of the thermoplastic liquid-crystal polymer films 6, 9 with lower heat resistance to a temperature that is 5°C higher than said melting point. As to the heating rolls 7, 10, metal rolls of, for example, induction heating mode or heat by medium oil circulating mode, are desirable from the viewpoint of uniform surface temperatures, and it is of no importance whether or not a coating layer is provided on the roll surface. For the coating layer on the surface, an elastic material such as rubber or a resin material with mold releasability such as a polyimide or Teflon can be used favorably. The diameter of each heating roll of the pair is preferably in the range of 35-45 cm, and it is preferred that both heating rolls of the pair have approximately the same diameter. Furthermore, the difference in the melting points of high-heat resistance thermoplastic liquid-crystal polymer film 1 and lower-heat resistance thermoplastic liquid-crystal polymer films 6, 9 is preferably 10°C or more, more preferably 25°C or more, and especially 50°C or more.

[0028] In the above-mentioned production methods, the pressure to be applied on respective thermoplastic liquid-crystal polymer films 1, 6, 9, and metal sheet 2 by heating rolls 3, 7, 10 is preferably 20 kg/cm<sup>2</sup> or more as surface pressure when using rolls that substantially deform at the pressing

parts. When using rolls that do not substantially deform at the pressing parts, pressures of 5 kg/cm or more as linear pressure are preferred in terms of producing sufficient adhesive strength. A production arrangement of this type can be used to produce sufficient adhesive strength while controlling the occurrence of variations. The upper limit of the pressure is not particularly critical, but in order to produce sufficient adhesive strength of the circuit substrate without flow and bulging of the circuit substrate during pressing of the lower-heat-resistance thermoplastic liquid-crystal polymer film, the pressure preferably does not exceed 200 kg/cm as linear pressure, or does not exceed 100 kg/cm<sup>2</sup> as surface pressure. When the surface temperature of the heating rolls is in the low temperature range, the flow and the bulging from the circuit substrate of the lower-heat-resistance thermoplastic liquid-crystal polymer film do not occur even if the above-mentioned pressure is exceeded.

[0029] Here, the linear pressure of the heating roll is the value of the force given to the heating roll (pressure bonding load) divided by the effective width of the heating roll. Furthermore, the surface pressure of the heating roll is the value of the pressure bonding load divided by the area of the pressing surface formed by deformation of the heating roll during pressure bonding.

[0030] To obtain a multilayer-circuit substrate with good appearance and excellent dimensional stability and adhesive strength by the above-mentioned inventions, it is desirable to adjust the rotational speed of the heating rolls to 30 m/minute or less as the linear speed of its circumference, when pressure bonding the high-heat-resistance circuit substrates and the lower-heat-resistance liquid-crystal polymer films by passing them between the heating rolls at temperatures between a temperature of 50°C lower than the melting point of the lower-heat-resistance liquid-crystal polymer film and a temperature of 5°C lower [sic; "higher" -- Tr. Ed.] than said melting point. In par-

ticular, the rotational speed is preferably adjusted to 20 m/minute or less, in order to facilitate heat transfer to the high-heat-resistance circuit substrates. The lower limit of the rotational speed of the heating rolls is not particularly critical, but it is industrially desirable not to reduce it to less than 0.1 m/minute, because rotational speeds that are too low cause major reductions in production efficiency.

[0031]

**[Actual Examples]** An actual example will now be presented to describe the present invention in concrete terms, but it is to be understood that this example is not intended to limit the present invention in any way. Furthermore, the melting point of a thermoplastic polymer film, the dimensional change in a multilayer-circuit substrate before and after pressure bonding with heating rolls, adhesive strength, etc., were measured and the appearance was evaluated as described in the following actual example and comparison example.

[0032]

(1) Melting Point

A differential scanning calorimeter was used, and the thermal behavior of a film was observed. In other words, a test specimen film was heated at a rate of 20°C/minute and melted completely, then the melt was cooled rapidly to 50°C at a rate of 50°C/minute and heated again at a rate of 20°C/minute, and the location of the endothermic peak that appeared in this heating was recorded as the melting point of the film.

[0033]

(2) Adhesive Strength

A peeling test specimen 1.0-cm wide was prepared from a multilayer-circuit substrate, its lower-heat-resistance film layer was affixed with two-sided adhesive tape to a flat plate, and the strength when the high-heat-

resistance circuit substrate layer was peeled away at a rate of 50 mm/minute by the 180° method in accordance with JIS C 5016 was measured.

[0034]

(3) Change in Dimension Before and After Heating Roll Pressure Bonding

Dimensional stability was measured in terms of a total of 9 [sic? 3 + 3 = 6? -- Tr. Ed.] points, 3 points in the length direction and 3 points in the width direction, in accordance with IPC-TM-650 2.2.4, and was given as a percent change based on the dimensions of the circuit substrate before pressure bonding by the heating rolls.

[0035]

(4) Appearance

Multilayer-circuit substrates were examined by visual inspection; those in which no "air grips" (these are observed as blisters) and no resin flows at all were observed over a length of 10 m were evaluated as good; those in which less than one air grip were observed per meter of length or resin flows of 1 mm or less were observed were evaluated as fair; and those where one or more air grips or non-sticking parts were observed per meter of length or resin flows of 1 mm or more were observed were evaluated as poor.

[0036]

(5) Thermal Expansion Coefficient

The amounts of thermal expansion between 30°C and 150°C of the thermoplastic liquid-crystal polymer films to be used in a circuit substrate and a multilayer-circuit substrate were measured with the use of a thermomechanical analyzer, and the thermal expansion coefficients were calculated from said amounts.

[0037]

Reference Example 1

A thermoplastic liquid-crystal polymer, which was a copolymer of p-

hydroxybenzoic acid and 6-hydroxy-2-naphthoic acid with a melting point of 280°C, was melt extruded at a discharge rate of 20 kg/hour, and formed into a film by inflation under the conditions of a transverse draw ratio of 4.77 and a longitudinal draw ratio of 2.09. This process produced a thermoplastic liquid-crystal polymer film with an average thickness of 50 µm and a small film thickness distribution, i.e., with a film thickness distribution of  $\pm$  7%. Subsequently, a heat treatment was carried out in a hot air dryer in a nitrogen atmosphere in order to increase the heat resistance. This heat treatment was carried out at 265°C for 1 hour and then at 275°C for 6 hours. As a result, a high-heat-resistance thermoplastic liquid-crystal polymer film with a melting point of 330°C was obtained. Furthermore, this high-heat-resistance liquid-crystal polymer film was placed between two metal sheets (electrolytic copper foil 18 µm thick), and these were pressure bonded and integrated into one piece between a pair of heating rolls with the surface temperature adjusted to 320°C, according to the method shown in Figure 1, to obtain a two-side, metal-lined laminated with high heat resistance. Subsequently, a circuit pattern to evaluate the change in the dimension before and after pressure bonding by the heating rolls was prepared on each metal sheet of the metal-lined laminate. The circuit pattern was chemically removed from the obtained metal-lined laminate, and the thermal expansion coefficient was measured, which was  $+5 \times 10^{-6}$  cm/cm/°C.

[0038]

Reference Example 2

A thermoplastic liquid-crystal polymer, which was a copolymer of *p*-hydroxybenzoic acid and 6-hydroxy-2-naphthoic acid with a melting point of 280°C, was melt extruded at a discharge rate of 20 kg/hour, and formed into a film by inflation under the conditions of a transverse draw ratio of 3.18 and a longitudinal draw ratio of 2.09. This process produced a thermoplastic liq-

uid-crystal polymer film with lower heat resistance with an average thickness of 75  $\mu\text{m}$  and a small film thickness distribution, i.e., with a film thickness distribution of  $\pm 7\%$ . The thermal expansion coefficient of the obtained film was  $-5 \times 10^{-6} \text{ cm/cm}/^\circ\text{C}$ .

[0039]

Actual Example 1

While feeding two pieces of the high-heat-resistance circuit substrates obtained in accordance with Reference Example 1 to the upper and lower surfaces of the lower-heat-resistance thermoplastic liquid-crystal polymer film obtained in accordance with Reference Example 2, the pieces were passed between a pair of first heating rolls, upper and lower, based on the method shown in Figure 2, whereby said circuit substrates and lower-heat-resistance thermoplastic liquid-crystal polymer film were pressure bonded and integrated into one piece. Subsequently, the lower-heat-resistance thermoplastic liquid-crystal polymer films were fed to the upper and lower surfaces of the above-mentioned laminate, and these were passed between a pair of second heating rolls, whereby a four-layer-circuit substrate with the pressure bonded lower-heat-resistance thermoplastic liquid-crystal polymer films on its upper and lower surfaces was prepared. The mold-release sheet placed around each heating roll in this case was a glass woven cloth-impregnated Teflon sheet, the surface temperature of each heating roll was adjusted to  $300^\circ\text{C}$  [*this is not consistent with the range given in "[0027]", i.e.,  $mp - 50^\circ\text{C} \leq T \leq mp + 5^\circ\text{C}$  -- Tr. Ed.*], and the surface pressure of each heating roll was adjusted to  $20 \text{ kg/cm}^2$ . the four-layer-circuit substrate obtained by removing the glass woven cloth-impregnated Teflon sheet continuously was suitable in appearance and had sufficient adhesive strength, i.e.,  $0.8 \text{ kg/cm}$  or more, with no variations in the width direction. The change in the dimensions before and after pressure bonding by the heating rolls did not vary with location and was satisfactory,

i.e., 0.1% or less at all locations.

[0040]

Comparison Example 1

Two pieces of the high-heat-resistance circuit substrates obtained in accordance with Reference Example 1 were laid over the upper and lower surfaces of the lower-heat-resistance thermoplastic liquid-crystal polymer films obtained in accordance with Reference Example 2, and moreover glass woven cloth-impregnated Teflon sheets were arranged above and below said stack; this assembly was placed on the hot plate of a vacuum hot press (flat-plate heat press) heated to 260°C, held in a vacuum condition for 15 minutes, then heated to 300°C, and held at a pressure of 20 kg/cm<sup>2</sup> for 10 minutes. The assembly was then cooled to 100°C, the pressure was dropped to atmospheric pressure, and the four-layer-circuit substrate was taken out. The four-layer-circuit substrate obtained had very noticeable resin flows in the circumference and its appearance was poor. The adhesive strength was 0.8 kg/cm around the center of the four-layer-circuit substrate, but was low, i.e., 0.5 kg/cm in the circumference, and moreover had variations. The change in the dimensions before and after heat and pressure bonding was 0.1% around the center, but was large, i.e., 0.3%, in the circumference, and there were also variations. Based on these results, the appearance was evaluated as poor.

[0041]

**[Effect of the Invention]** As described above, the present invention can be used for the production of multilayer-circuit substrates with good appearance, sufficient dimensional stability, and high adhesive strength at low cost.

**[Brief Description of the Figures]**

**[Figure 1]** Figure 1 is a diagram that schematically illustrates one example of a method for producing a high-heat-resistance metal-lined laminate to

be used in the present invention.

[Figure 2] Figure 2 is a diagram that schematically illustrates one example of the method for producing a multilayer-circuit substrate in accordance with the present invention.

**[Description of the Symbols]**

(1) thermoplastic liquid-crystal polymer film with high heat resistance; (2) metal sheet; (3) heating roll; (4) two-side metal-lined laminate; (5) circuit substrate; (6, 9) thermoplastic liquid-crystal polymer film with lower heat resistance; (7) first heating roll; (10) second heating roll; and (A, B) multilayer-circuit substrate.

**[Figure 1]**

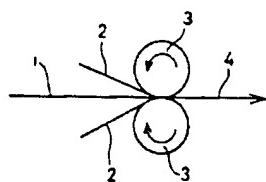


Figure 1. (1) thermoplastic liquid-crystal polymer film with high heat resistance; (2) metal sheet; (3) heating roll; and (4) two-side metal-lined laminate.

**[Figure 2]**

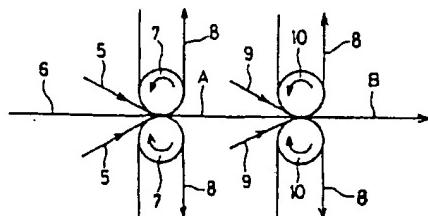


Figure 2. (5) circuit substrate; (6, 9) thermoplastic liquid-crystal polymer film with lower heat resistance; (7) first heating roll; (10) second heating roll; and (A, B) multilayer circuit substrate.